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## Biogenic versus anthropogenic sources of CO in the United States

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[1] Aircraft observations of carbon monoxide (CO) from the ICARTT campaign over the eastern United States in summer 2004 (July 1–August 15), interpreted with a global 3-D model of tropospheric chemistry (GEOS-Chem), show that the national anthropogenic emission inventory from the U.S. Environmental Protection Agency (93 Tg CO y<sup>-1</sup>) is too high by 60% in summer. Our best estimate of the CO anthropogenic source for the ICARTT period is 6.4 Tg CO, including 4.6 Tg from direct emission and 1.8 Tg CO from oxidation of anthropogenic volatile organic compounds (VOCs). The biogenic CO source for the same period from the oxidation of isoprene and other biogenic VOCs is 8.3 Tg CO, and is independently constrained by ICARTT observations of formaldehyde (HCHO). Anthropogenic emissions of CO in the U.S. have decreased to the point that they are now lower than the biogenic source in summer.

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### 1. Introduction

[2] Carbon monoxide (CO) is emitted to the atmosphere by combustion, and is also produced within the atmosphere by oxidation of volatile organic compounds (VOCs). Anthropogenic CO emissions presently contribute a  $\pm 0.20$  W m<sup>-2</sup> radiative forcing of climate change according to *Intergovernmental Panel on Climate Change* [2007], mainly through chemical effects on the concentrations of tropospheric ozone and methane. CO is also a toxic gas at high concentrations and hence the subject of regulation. The U.S. Environmental Protection Agency (EPA) (Current emissions trends summaries from

the NEI, 2007, available at <http://www.epa.gov/ttn/chief/trends/>) (hereinafter referred to as EPA online report, 2007) claims that U.S. anthropogenic emissions of CO have been decreasing on average by 2.6% y<sup>-1</sup> since 1990, but there is large uncertainty in the EPA inventory. Observations in urban air suggest that EPA estimates of on-road vehicular emissions, accounting for 2/3 of the detailed 1999 EPA National Emission Inventory (NEI 99) for CO, are 50% too high [Parrish, 2006; Warneke *et al.*, 2006]. We use here boundary layer observations from the ICARTT aircraft campaign over the eastern U.S. in summer 2004 to better quantify the U.S. anthropogenic source of CO and compare it to the natural source from oxidation of biogenic VOCs emitted by vegetation. As we will see, the anthropogenic source has decreased to the point that the biogenic source now dominates in summer.

### 2. ICARTT Aircraft Campaign

[3] ICARTT was a coordinated multi-aircraft atmospheric chemistry field program over eastern North America and the North Atlantic in July–August 2004. The NOAA WP-3D aircraft [Fehsenfeld *et al.*, 2006] and the DC-8 aircraft [Singh *et al.*, 2006] flew extensive boundary layers legs across the eastern U.S. over the July 1–August 15 period, typically at 300 m over land. The two aircraft showed agreed in their CO measurement to within  $\pm 5\%$  (G. Chen, personal communication, 2005; <http://www-air.larc.nasa.gov/missions/intexna/meas-comparison.htm>) and are used here as a single data set. Other aircraft measurements used in this paper include propane, nitrogen oxide radicals, reactive nitrogen oxides (NO<sub>y</sub>), propane, HCN, and CH<sub>3</sub>CN. Surface measurements of CO were also taken during ICARTT from Chebogue Point on the southern tip of Nova Scotia (44°N, 66°W) to observe outflow from the northeastern U.S. [Fehsenfeld *et al.*, 2006; Millet *et al.*, 2006a]. See above references for instrument details.

### 3. Model Description

[4] We simulate the ICARTT observations with the GEOS-Chem global 3-D model of tropospheric chemistry (version 7.02; <http://www.as.harvard.edu/chemistry/trop/geos/>) driven by assimilated meteorological observations from the Goddard Earth Observing System (GEOS-4) of the NASA Global Modeling and Assimilation Office (GMAO). The model is applied to a global simulation of ozone-NO<sub>x</sub>-VOC-aerosol chemistry. A general description of GEOS-Chem is given by Bey *et al.* [2001] and a specific description of the coupled oxidant-aerosol simulation as used here is given by Hudman *et al.* [2007]. The horizontal resolution is 2° × 2.5°. There are 30 vertical layers including 12 below 10 km and 5 below 2 km. The

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**Table 1.** CO Sources Over the Contiguous United States for 1 July to 15 August 2004<sup>a</sup>

Source Type	Magnitude, Tg CO
Fuel Combustion	4.6 <sup>b</sup>
Oxidation of anthropogenic VOCs	1.8
Oxidation of Biogenic VOCs	8.3 ± 2.5 <sup>c</sup>
Biomass Burning	0.2 <sup>d</sup>
Total	14.9

<sup>a</sup>Not including the oxidation of methane and minor long-lived VOCs (such as methanol and acetone) which provide a background source throughout the troposphere (this source is included in the GEOS-Chem simulation).

<sup>b</sup>Constrained by the ICARTT observations as discussed in the text. This is 60% lower than the source of 11.5 Tg given by the EPA NEI 99.

<sup>c</sup>Including 6.7 Tg from isoprene oxidation and 1.6 Tg from oxidation of other biogenic VOCs (monoterpenes,  $\geq$ C3 alkenes). The 30% uncertainty is constrained by ICARTT observations of HCHO as discussed in the text.

<sup>d</sup>North American fires during ICARTT were mainly outside the contiguous United States; Alaskan and Canadian fires emitted 19 Tg CO during this period [Turquety *et al.*, 2007].

simulation is spun up for 12 months starting from chemical climatology; this effectively removes the influence of initial conditions.

[5] For comparison with observations, the model is sampled along the aircraft flight tracks for the flight times. We focus here on observations in the boundary layer below 1.5 km altitude, and exclude fresh pollution plumes because they cannot be accurately represented at the coarse resolution of the model. We diagnose fresh pollution plumes in the observations by  $\text{NO}_x/\text{NO}_y > 0.4 \text{ mol mol}^{-1}$  or (if  $\text{NO}_y$  is not available)  $\text{NO}_2 > 4 \text{ ppbv}$ . We also remove biomass burning plumes as diagnosed by  $\text{HCN} > 500 \text{ pptv}$  or  $\text{CH}_3\text{CN} > 225 \text{ pptv}$  [de Gouw *et al.*, 2006]. Several of the WP-3D flights were targeted at urban plume characterization [Fehsenfeld *et al.*, 2006] and we do not use those data. These filters exclude 7% and 21% of the DC-8 and WP-3D data, respectively.

[6] Global anthropogenic emissions in the model are as described by Park *et al.* [2004]. Sources of CO for the United States during ICARTT (July 1–August 15, 2004) are summarized in Table 1 and vary by month, weekday/weekend, and time of day. The initial simulation used an anthropogenic fuel source of 13.3 Tg CO from NEI 99, including 1.8 Tg CO from oxidation of anthropogenic VOCs. This source was subsequently decreased by 60% to 6.4 Tg CO on the basis of the ICARTT observational constraints, as discussed below. The biogenic VOC source of CO is 8.3 Tg CO, including 6.7 Tg CO from isoprene and 1.6 Tg CO from other short-lived biogenic VOCs including monoterpenes and alkenes. Emissions of isoprene and monoterpenes are from the GEIA inventory and are computed locally as a function of solar radiation and temperature [Guenther *et al.*, 1995]. The CO yields from VOC oxidation are calculated for the purpose of Table 1 using yield data from Duncan *et al.* [2007], but are actually simulated in GEOS-Chem. For isoprene we use in Table 1 a yield of 0.45 per C atom, consistent with the GEOS-Chem mechanism [Palmer *et al.*, 2003] and with formaldehyde observations during ICARTT [Millet *et al.*, 2006a]. Wildfire emissions are from Turquety *et al.* [2007] and are negligibly

low for the contiguous U.S. (0.2 Tg CO). However, large fires occurred over Alaska and western Canada during ICARTT.

#### 4. ICARTT Constraints on CO Sources

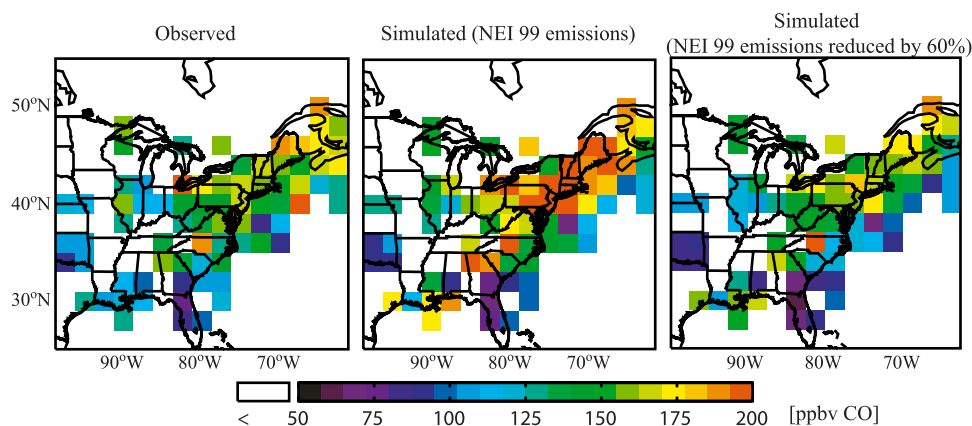
[7] Figure 1 compares simulated and observed mean CO concentrations in the boundary layer (0–1.5 km altitude) during ICARTT. The model using NEI 99 emissions shows a consistent 20–50 ppbv overestimate across eastern North America that matches the spatial distribution of the anthropogenic CO source. Surface observations at Chebogue Point show a similar overestimate (Figure 2).

[8] The model overestimate of CO reflects either excessive sources or insufficient boundary layer ventilation. A strong argument against the latter is that the model reproduces successfully the observed shapes of the mean vertical profiles for propane (auxiliary material Figure S1<sup>1</sup>), acetylene [Xiao *et al.*, 2007], and formaldehyde [Millet *et al.*, 2006b] during ICARTT. Chemical loss of CO from oxidation by OH is slow relative to boundary layer ventilation and is therefore of little consequence.

[9] The chemical source of CO in the model from VOC oxidation is well constrained by the successful simulation of formaldehyde (HCHO) observations aboard the DC-8 aircraft [Millet *et al.*, 2006b]. The entire chemical source of CO from VOC atmospheric oxidation, according to current understanding, passes through HCHO as an intermediate. Most of the boundary layer HCHO during ICARTT was from isoprene oxidation [Millet *et al.*, 2006b], as previously observed in eastern North America [Shepson *et al.*, 1991; Lee *et al.*, 1998]. Millet *et al.* [2006b] shows that GEOS-Chem, including the same VOC sources as here, simulates the DC-8 HCHO observations by A. Fried *et al.* (Formaldehyde over North America and the North Atlantic during the Summer 2004 INTEX campaign: Methods, observed distributions, and measurement box model comparisons, submitted to *Journal of Geophysical Research*, 2008) (hereinafter referred to as Fried *et al.*, submitted manuscript, 2008) with no significant bias. Co-located HCHO observations by Snow *et al.* [2007], aboard the DC-8, were 30% lower than Fried *et al.* (submitted manuscript, 2008) but highly correlated, and we use this difference as an estimate of the possible error in our biogenic CO source estimate (Table 1).

[10] It thus appears that the model bias in Figure 1 is due to an excessive fuel combustion source in the NEI 99 inventory, and this is consistent with the spatial structure of the bias. Chemical production from oxidation of anthropogenic VOCs is only 14% of that source and is probably not overestimated since the model shows no bias for propane during ICARTT (auxiliary material Figure S1) and is too low for ethane and acetylene [Xiao *et al.*, 2007, also Global budget of ethane and constraints on North American sources from INTEX-A aircraft data, manuscript in preparation, 2008]. We conclude that CO anthropogenic emission is greatly overestimated in the NEI 99 inventory.

<sup>1</sup>Auxiliary materials are available in the HTML. doi:10.1029/2007GL032393.



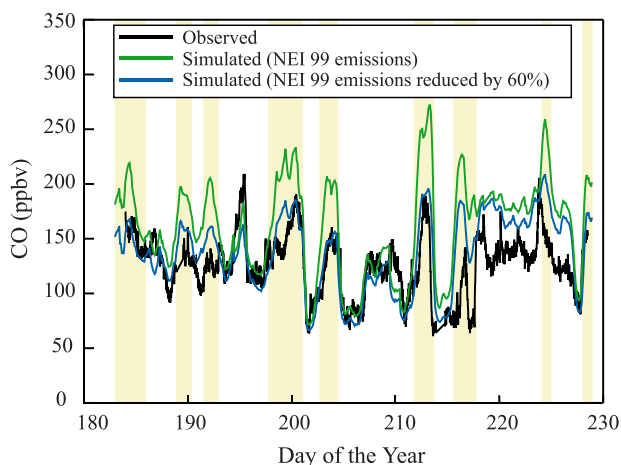
**Figure 1.** Mean CO concentrations in the boundary layer (0–1.5 km altitude) during ICARTT (July 1–August 15, 2004). (left) Observations averaged over the  $2^\circ \times 2.5^\circ$  GEOS-Chem model grid are compared to model results using the (middle) U.S. EPA NEI 99 emissions and (right) anthropogenic CO emissions reduced by 60%. Model results are sampled along the flight tracks at the time of the flights.

[11] Figure 3 shows the correlation of simulated versus observed CO concentrations at 0–1.5 km altitude. Here and elsewhere, regression lines are derived from the reduced-major-axis (RMA) method allowing for errors on both variables, and 95% confidence intervals are determined by bootstrap re-sampling ( $n = \text{sample size}$ ). The regression line indicates an overestimate of  $32 \pm 15\%$ . This result is insensitive to the exclusion of fresh pollution plumes from the comparison. We find that correcting the bias requires reducing anthropogenic CO emissions during the ICARTT period from 11.5 to 4.6 Tg CO, i.e., a 60% decrease relative to the NEI 99 values (Figure 3, bottom). Applying this decrease also improves the simulated geographical distribution (Figure 1), and the simulation at Chebogue Point (Figure 2). A 60% decrease in the anthropogenic emission source relative to NEI 99 is consistent with the 50% biases reported by Parrish [2006] and Warneke *et al.* [2006] for urban air dominated by on-road vehicles. It indicates that the bias extends over the Eastern U.S. and that other anthropogenic CO sources from off-road vehicles and industry must be overestimated as well. Assuming that the 60% downward correction applies year-round, and to the entire U.S., our best estimate of the U.S. national emission in 2004 is 37 Tg  $\text{y}^{-1}$ .

[12] After correction of this bias, we find in Table 1 that anthropogenic CO emissions account for only 31% of the total U.S. source of CO in summer. Anthropogenic VOC emissions contribute an additional 12%, making the anthropogenic contribution 43%. Biogenic VOC emissions contribute 56%. Isoprene is the largest biogenic source of CO, contributing 45% of the total CO source. Other biogenic VOCs contribute 11%. This is even though the summer of 2004 was unusually cool [Fuelberg *et al.*, 2007]. In a warmer year the contribution from biogenic VOCs would be even higher.

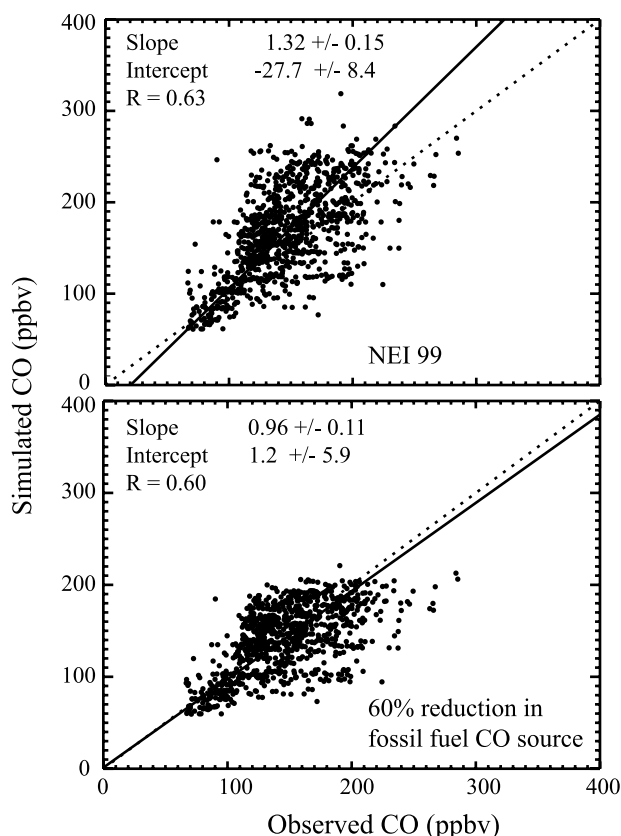
[13] We compare in Figure 4 the simulated CO concentration enhancements from U.S. anthropogenic and biogenic sources, as determined by difference between the standard simulation and sensitivity simulations with these sources shut off. Biogenic CO enhancements are highest in the

Southeast where isoprene emissions are highest, while anthropogenic CO enhancements are highest in the Northeast where combustion sources are highest. The mean enhancements over eastern North America are the same in the two cases, even though the biogenic source is larger. This is because isoprene suppresses OH concentrations in current photochemical model mechanisms [Jacob and Wofsy, 1988] (also X. R. Ren *et al.*, HOx Chemistry during INTEX-A 2004: Observation, model calculations, and comparison with previous studies, submitted to *Journal of Geophysical Research*, 2008) (hereinafter referred to as Ren *et al.*, submitted manuscript, 2008), so that shutting off isoprene emission increases the boundary layer CO source from methane oxidation. Aircraft observations of OH during ICARTT do not show a depletion of OH



**Figure 2.** Surface air CO concentrations at Chebogue Point during ICARTT. Observations (black) are compared to model results using NEI 99 anthropogenic emissions (green) and with these CO emissions reduced by 60% (blue). Yellow bands are periods of U.S. outflow diagnosed by Millet *et al.* [2006b]. Overestimate near day 220 is due model misplacement of a large Alaskan/Canadian biomass burning plume.





**Figure 3.** Scatterplot of simulated versus observed CO concentrations at 0–1.5 km altitude during ICARTT. Model results are from the simulation using NEI 99 anthropogenic CO emissions (top) and the simulation with these emissions reduced by 60% (bottom). Reduced-major-axis regressions (black) and  $y = x$  lines (dotted black) are shown. The Pearson correlation coefficient and the regression line parameters (with  $\pm$  95% confidence interval calculated by the bootstrap method) are given as legends.

correlated with isoprene, pointing to an error in these mechanisms (Ren et al., submitted manuscript, 2008). This problem requires further investigation and implies that the

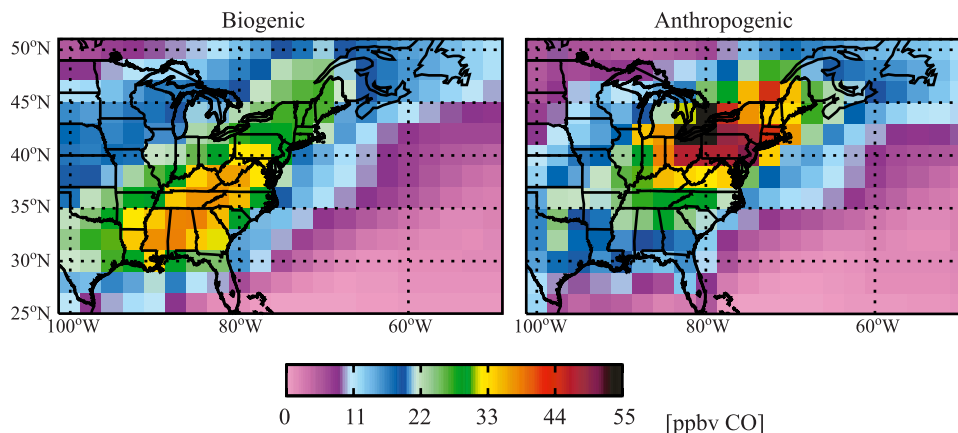
CO enhancement from biogenic VOCs is greater than shown in Figure 4.

## 5. Conclusions

[14] Application of the GEOS-Chem model to simulate ICARTT aircraft observations for CO over eastern North America in July–August 2004 shows that U.S. anthropogenic CO emissions are 60% lower than the U.S. EPA inventory for 1999 (NEI 99). EPA (online report, 2007) reported a mean  $2.6\% \text{ y}^{-1}$  decrease of CO emissions over the 1990–2004 period; this would imply a 12% decrease for 1999–2004, insufficient to account for the bias. Parrish [2006] and Warneke et al. [2006] found from analysis of urban CO data that on-road sources in the NEI 99 inventory are 50% too high. Our analysis implies that other anthropogenic CO sources from off-road vehicles and industry must also be overestimated.

[15] Our resulting best estimate of U.S. anthropogenic CO emissions for the ICARTT period (July 1–August 15, 2004) is 4.6 Tg, which combined with our best estimate of the secondary source from oxidation of anthropogenic VOCs (1.8 Tg) yields a total anthropogenic source for the period of 6.4 Tg. This can be compared to the CO source from oxidation of biogenic VOCs, which is well constrained during the ICARTT period by the successful GEOS-Chem simulation of aircraft observations of HCHO [Millet et al., 2006b]. We find a biogenic source of CO for the ICARTT period of 8.6 Tg (6.7 Tg from isoprene, 1.6 Tg from other VOCs). The anthropogenic CO source in the United States is thus lower than the biogenic source during summer.

[16] The simulated enhancement of CO concentrations from biogenic sources in the model is comparable to that from anthropogenic sources and weaker than one would expect on the basis of biogenic VOC emissions. This is because of model suppression of OH when isoprene concentrations are high. Observations of OH during ICARTT do not show such isoprene-driven OH titration. A possible reason could be the presence of a large biogenic OH source from ozonolysis of biogenic VOCs missing from current models [Goldstein et al., 2004; Farmer and Cohen, 2007; Kuhn et al., 2007]. This model bias mandates further investigation but it does not affect the constraints on CO sources derived here.



**Figure 4.** July mean enhancements of CO concentrations at 0–2.5 km altitude from (left) anthropogenic and (right) biogenic North American sources, as determined by difference between the standard simulation and simulations with these sources shut off in the domain ( $130^{\circ}$ – $70^{\circ}$ W,  $25^{\circ}$ – $50^{\circ}$ N).

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